

Particulate structure of FePt thin films enhanced by Au and Ag alloying

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The additions of Au and Ag to FePt thin film change the film structure from an interconnected network to a completely isolated island structure. In particular, the Ag addition led to an isolated particulate structure smaller than 100 nm, which increased the coercivity from 5.4 to 20.5 kOe. Energy filtered elemental mapping did not show evidence for the precipitation or segregation of Au and Ag. © 2006 American Institute of Physics. [DOI: 10.1063/1.2335600]

To overcome the thermal instability of nanosized ferromagnetic particles, $L1_0$ ordered FePt phase with a high magnetocrystalline anisotropy of about 7×10^7 ergs/cc is considered to be promising for future ultrahigh density recording media. Magnetically decoupled $L1_0$ -FePt nanoparticles must be aligned with the c axis normal to the film plane to be utilized as perpendicular recording media.¹ However, the FePt particle assemblies that are fabricated by either sputtering or chemical synthesis usually have disordered fcc ($A1$) structures, which are either superparamagnetic or soft magnetic. The phase transformation from $A1$ to $L1_0$ is necessary to achieve the hard magnetic properties that are suitable for recording media applications.^{2,3} In addition, particle isolation and c -axis alignment must be achieved to prepare the FePt thin films for ultrahigh density perpendicular recording media.

Many previous investigations reported the effect of Au and Ag additions in FePt thin films.⁴⁻⁷ In particular, the Ag addition was reported to decrease the ordering temperature of FePt thin films effectively, which is attributed to the introduction of vacancies during the rejection of immiscible Ag atoms from the FePt nanoparticles.^{4,5,7} The promotion of the ordering was also reported for the FePt film with Ag top layer,⁸ with Ag underlayer,⁹ and FePt/Ag multilayer film,¹⁰ in which case the rejection of Ag from the FePt layer was not expected. Although the rejection of Ag from the FePt was reported in the earlier work,⁷ recent investigation has shown that Ag dissolves in the $L1_0$ FePt phase.¹¹

Sputter deposition of FePt on heated substrates at around 700 °C leads to the formation of a particulate film of fully ordered $L1_0$ FePt by the Volmer-Weber growth mode.¹² However, due to the sluggish atomic mobility of FePt, the continuous film structures are always obtained at temperatures below 550 °C.¹³ A high substrate temperature is needed to achieve the particulate structure of an ordered FePt with high coercivity, which is not favorable for practical applications due to the limited temperature tolerance of recording media substrates.^{12,13} In addition, such high temperature process causes rapid growth of particles, which results in disappointing bimodal distribution of particles with particle size as large as 100 nm.¹⁴ Alloying FePt with some elements that have a lower melting temperature may alter the growth mode

at a lower temperature and may lead to reduction of the island grown particle size. In this work, c -axis perpendicular alignment was realized by using the epitaxial growth on the MgO (001) single crystal substrate. The formation of a particulate structure and an ordering process were enhanced by alloying FePt with Au and Ag that have lower melting temperatures, resulting in high coercive perpendicular anisotropy film at 520 °C.

FePt, (FePt)₉₀Au₁₀, and (FePt)₉₀Ag₁₀ films were prepared by cosputtering high-purity ~99.99% Fe, Pt, Au, and Ag targets on heated MgO (001) single crystal substrates using a multiple dc-sputtering system. The base pressure of the system was less than 1×10^{-6} Pa and the high-purity argon of ~0.1 Pa was flown during sputtering. The nominal film thickness was fixed to 10 nm based on the sputtering time. The substrate temperature (T_s) was kept at 520 °C on a rotating table during film deposition. Magnetization curves were measured normal to the film plane using a superconducting quantum interference device (SQUID) magnetometer with a maximum magnetic field of 5.5 T. The film orientation and the degree of ordering were evaluated by x-ray diffraction (XRD). The plane view microstructures of the films were observed by Philips CM 200 and JEOL 2010F transmission electron microscopes (TEMs).

Figure 1 shows the XRD patterns of the FePt, (FePt)₉₀Au₁₀, and (FePt)₉₀Ag₁₀ films that were deposited on the MgO (100) substrate at 520 °C. All the peaks can be indexed as an ordered $L1_0$ FePt except for the unindexed peaks of the substrate. The subpeaks on the left side of the (001) peaks of the Au and Ag containing films originate from the substrate. No additional peaks from Au and Ag are found, which agrees with the previous work in which the Ag diffraction peak could only be observed when the content of Ag was higher than 20 at. % and the postannealing temperature was higher than 550 °C.⁵ The strong (001) superlattice reflection from the FePt film indicates that the film has the $L1_0$ structure from the as-deposited state. Only the (00 n) diffractions are observed in all the films due to the epitaxial growth of the (001) FePt on the MgO (001) substrate. Figure 1 shows that the intensity of the (001) superlattice reflection becomes stronger with Au and Ag alloying. The degree of order S was evaluated from the integrated intensities of the (002) fundamental peak and the (001) superlattice peak using $S^2 = (I_{\text{super}}/I_{\text{fund}})_{\text{exp}} / (I_{\text{super}}/I_{\text{fund}})_{\text{theo}}^{S=1}$. The theoretical intensities

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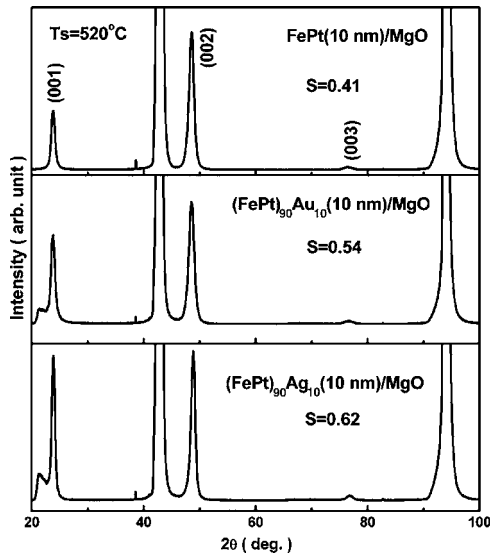


FIG. 1. XRD patterns of FePt, $(\text{FePt})_{90}\text{Au}_{10}$, and $(\text{FePt})_{90}\text{Ag}_{10}$ films deposited on a heated MgO substrate at 520°C .

of both the fundamental and superlattice diffractions were estimated by considering the following parameters: atomic fractions, atomic scattering factors, Debye-Waller corrections, Lorentz polarization factors, and structure factors. The values of degree of order were 0.41, 0.54, and 0.62 for FePt, $(\text{FePt})_{90}\text{Au}_{10}$, and $(\text{FePt})_{90}\text{Ag}_{10}$, respectively, showing an enhancement of ordering by alloying FePt with Au and Ag. The film with the Ag addition exhibits the highest degree of order.

Figure 2 shows the magnetization curves of the three films measured along the film plane and normal to the film plane. The estimated values of anisotropy field (H_k) and magnetocrystalline anisotropy (K_u) are shown in the figures. The saturation magnetization decreased as a result of Au and Ag alloying. The values of the anisotropy field (H_k) and the magnetocrystalline anisotropy (K_u) increased by alloying FePt with Au and Ag. If the degree of order is the same, both H_k and K_u should be the highest for FePt binary alloy, while $(\text{FePt})_{90}\text{Au}_{10}$ and $(\text{FePt})_{90}\text{Ag}_{10}$ show higher H_k and K_u . This suggests that the degree of $L1_0$ order is higher in the sample alloyed with Au and Ag. Since the $(\text{FePt})_{90}\text{Ag}_{10}$ sample shows the highest H_k and K_u values among three samples, we can conclude that Ag enhance the ordering most effectively with this deposition condition ($T_s=520^\circ\text{C}$). The coercivity was significantly enhanced from 5.4 to 9.4 kOe by adding Au. The coercivity increased drastically to 20.5 kOe for the film with the Ag addition, which is similar to that reported for the *in situ* annealed FePt thin films with Ag top layer by Zhao *et al.*⁸ The increases of magnetic anisotropic field (H_k) or anisotropy coefficient (K_u) cannot explain such significant enhancement of the coercivities alone, indicating that microstructural modification by Au and Ag additions also plays a critical role in coercivities. We found large changes in the morphology of the films by the addition of Au and Ag as shown in Fig. 3, which is thought to be more influential on the coercivity change.

Figure 3 shows in-plane TEM images and their selected area electron diffraction (SAED) patterns of the three types

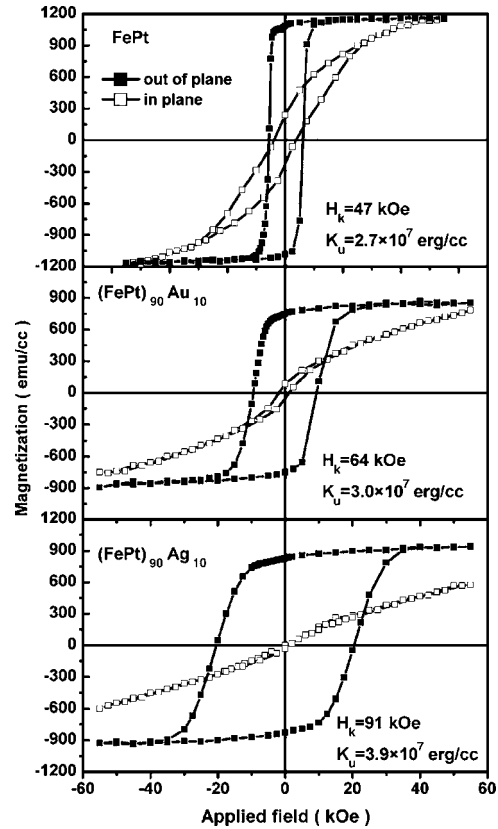


FIG. 2. In-plane and out-of-plane magnetization curves of the FePt, $(\text{FePt})_{90}\text{Au}_{10}$, and $(\text{FePt})_{90}\text{Ag}_{10}$ films. Anisotropy field (H_k) and magnetocrystalline anisotropy constant (K_u) were estimated by extrapolating the in-plane and out-of-plane magnetization curves.

of films. The SAED patterns confirm the epitaxial growth of the FePt layer with the orientation relationship of $(001)\text{Pt}/(001)\text{MgO}$ and $[001]\text{FePt}/[001]\text{MgO}$. The appearance of only the $\{110\}$ superlattice spots suggests that the c axis is

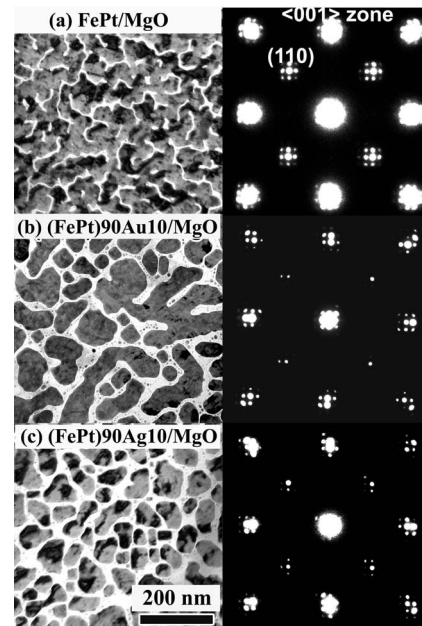


FIG. 3. Plane view TEM images and selected area diffraction patterns of FePt, $(\text{FePt})_{90}\text{Au}_{10}$, and $(\text{FePt})_{90}\text{Ag}_{10}$ films.

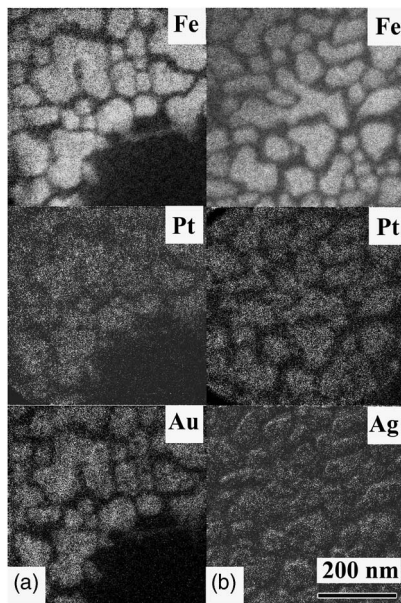


FIG. 4. Energy filtered elemental mappings of $(\text{FePt})_{90}\text{Au}_{10}$ (a) and $(\text{FePt})_{90}\text{Ag}_{10}$ (b) films.

perfectly oriented in the perpendicular direction to the film plane, which agrees with the XRD results. Although all of the films have nominally the same thickness, the morphologies of the films are very different. The FePt film is interconnected, but both the $(\text{FePt})_{90}\text{Au}_{10}$ and $(\text{FePt})_{90}\text{Ag}_{10}$ films show an isolated particulate microstructure. Since the FePt islands are interconnected, the magnetic reversal can occur easily by the nucleation of reverse magnetic domains and their wall displacement with little pinning force, resulting in low coercivity.^{11,15} Figure 3(b) shows that the morphology of the film becomes discontinuous by the addition of 10 at. % Au. However, some very large interconnected parts still remain. A completely isolated particulate structure was formed in the thin film with the 10 at. % Ag addition, as shown in Fig. 3(c). The size of almost all the particles is smaller than 100 nm with a small fraction of much smaller particles around 30 nm. Therefore, the difference in H_c of the $(\text{FePt})_{90}\text{Ag}_{10}$ and $(\text{FePt})_{90}\text{Au}_{10}$ particulate films is thought to be partly due to the difference of the isolated particle size, i.e., the possibility of the nucleation of reverse domains becomes smaller for smaller particles.¹³ In the FePt thin film, there are only a few pinning sites due to the lack of grain boundaries and lattice defects such as twins,¹⁶ so the interconnected microstructure causes low coercivity.¹²

To understand the effect of Au and Ag additions, the energy filtered TEM was applied to check the distributions of Au and Ag. The Gatan imaging filter (GIF) elemental mappings of the Au and Ag containing films are shown in Fig. 4. The Fe and Pt mappings of both samples show uniform intensity within the particles. The Au mapping of Fig. 4(a) is also uniform and indicates that Au does not precipitate to form elemental Au. Since the Ag edge does not appear clearly in the electron energy loss spectrum (EELS), the Ag mapping looks noisy; however, the uniformity of the Fe and Pt mappings suggests that Ag does not form elemental particles. These results are consistent with the XRD results of

no corresponding diffractions from Au and Ag. Although these results do not rule out the possibility of the surface segregation of Au and Ag, it can be concluded that Au and Ag do not form any elemental particles, thus they are most likely to be dissolved in FePt phase. This would influence the intrinsic magnetic properties such as saturation magnetization and magnetocrystalline anisotropy of FePt, but their details are beyond the scope of this paper.

The melting temperature of FePt is higher than 1500 °C, while those for Au and Ag are 1064 and 961 °C, respectively. The melting points of FePt alloy should be decreased by alloying with Au and Ag, resulting in the enhancement of atomic mobility compared to the binary system. By accelerating the atomic diffusions, the ordering processes are promoted and the formation of particulate structures are thought to be enhanced. The coercivity increase by the addition of Au and Ag should be mainly because of the morphology change of the film and the enhancement of the degree of order. The formation of perfectly isolated particles at reduced temperature in the Ag containing FePt thin film will be useful for designing FePt particulate films for potential recording media applications.

In summary, the isolation of magnetic particles and the promotion of ordering were realized at $T_s=520$ °C by alloying FePt with 10 at. % Au and Ag. An interconnected network structure in the binary 10 nm thick FePt thin film became particulate after alloying with 10 at. % Au and Ag at $T_s=520$ °C. In particular, the Ag containing film showed a completely isolated island structure with an island size smaller than 100 nm, resulting in a huge coercivity of 20.5 kOe.

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